Scaling investigation of the magnetic entropy change in helimagnet MnSi

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(Dated: July 14, 2015)

Abstract

The B20 compound MnSi exhibits a skyrmion phase in a certain range of temperature (T) and external magnetic field (H) in addition to the helimagnetic ordering. In this work, the magnetic entropy change (ΔS_M) in the single crystal helimagnet MnSi has been investigated by the scaling analysis. The $\Delta S_M(T, H)$ curves in higher field range (2 kOe $< H <$ 5.6 kOe) collapse onto an universal curve after scaling, which is independent of T and H. Based on this universal curve, the $\Delta S_M(T, H)$ (excluding the skyrmion entropy change) at any T and H can be calculated. On the other hand, a small valley is found on the $\Delta S_M(T)$ curve at ~ 28.5 K in the field range from ~ 1 kOe to ~ 2 kOe, which is resulted from the skyrmion phase. Subsequently, the magnetic entropy change of the skyrmion phase [$\Delta S_M^{skX}(T, H)$] can be derived by subtraction of the calculated $\Delta S_M^{cal}(T, H)$ from the experimental $\Delta S_M^{exp}(T, H)$. The obtained results show that ΔS_M^{skX} reaches the maximum at ~ 1.6 kOe, which indicates the highest number of skyrmion vortices at that field.

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I. INTRODUCTION

Recently, the skyrmion ordering has triggered great interest because of its great potential application in spintronics storage device due to the topological property [1, 2], nanometric size [3], and current-driven motion [4, 5, 6]. The skyrmion phase is a topologically stable spin vortex-like texture existing in a certain range of temperature and magnetic field [7]. The compounds exhibiting a skyrmion phase, such as FeGe [8], MnSi [9], $Fe_{1-x}Co_xSi$ [10, 11], Cu_2OSeO_3 [12, 13], etc, have been extensively and intensively investigated. Especially, much interest has been drawn on the B20 compound MnSi due to the additional weak itinerant ferromagnetism [14], non-Fermi-liquid behavior [15, 16], and magnetic quantum phase transition [17]. The cell of MnSi has cubic symmetry with space group $P2_13$. The lack of centrosymmetry results in a weak Dzyaloshinskii-Moriya (DM) spin-orbit coupling. The DM spin-orbit coupling competes with the much stronger ferromagnetic exchange, which finally causes a long modulation period of a helimagnetic ground state [1, 2, 18]. It has been demonstrated that MnSi exhibits a first-order phase transition into the helimagnetic ordering at 30.5 K, which can be suppressed into a second one by an external field where a tricritical point was induced [19, 20, 21, 22, 23]. When an external magnetic field is applied above a threshold value, the helimagnetic state transforms into a conical phase [7]. The chiral skyrmion phase emerges just below T_C in a narrow magnetic field range due to a unique stabilization provided by the DM interactions [1]. In the bulk material of MnSi, the skyrmion phase emerges at ~ 28 K in the field range from 1.55 kOe to 2.3 kOe [23]. It has been proved that the skyrmion phase can be stabilized by reducing the dimension of the materials [24, 25].

As we know, a magnetic ordering phase transition will result into a change of the magnetic entropy. Recently, a scaling analysis of the magnetic entropy change correlated to critical exponents has been proved an effective method to study the magnetic entropy change [26, 27, 28, 29]. In this work, the magnetic entropy change (ΔS_M) in the single crystal helimagnet MnSi has been investigated by the scaling analysis. The $\Delta S_M(T)$ curves exhibit a valley at 28.5 K in the magnetic range from ~ 1 kOe to ~ 2 kOe, which just corresponds to the formation of the skyrmion phase. The $\Delta S_M(T, H)$ curves for higher fields collapse onto an universal curve after scaling, which is independent of the temperature T and the applied external field H. Based on this universal curve, $\Delta S_M(T, H)$ (excluding the skyrmion entropy

change) at any T and H can be calculated. Subsequently, the magnetic entropy change of the skyrmion phase $[\Delta S_M^{skX}(T, H)]$ can be derived by subtraction of the calculated $\Delta S_M^{cal}(T, H)$ from the experimental $\Delta S_M^{exp}(T, H)$. It is found that ΔS_M^{skX} reaches the maximum at $\sim$ 1.6 kOe, which indicates the highest number of skyrmion vortices in MnSi at this magnetic field.

II. EXPERIMENT

A single crystal sample of MnSi was prepared by the Czochralski method [30]. The specimen quality and physical properties were checked and described elsewhere [22]. The magnetization was measured using a Quantum Design vibrating sample magnetometer (SQUID-VSM). The no-overshoot mode was applied to ensure a precise magnetic field. The applied field was relaxed for 2 minutes before the data collection. For the measurement the sample was processed into slender ellipsoid shape, and the magnetic field was applied along the longest axis to minimize the demagnetizing field. To make sure that each curve was initially magnetized, the isothermal magnetization measurement was performed after the sample was heated well above T_C for 10 minutes and then cooled under zero field to the target temperatures. The magnetic background was carefully subtracted. The applied magnetic field H_a has been corrected into the internal field as $H = H_a - NM$ (where M is the measured magnetization and N is the demagnetization factor) [31]. The corrected H was used for the calculation of the magnetic entropy change. All the magnetic units and formulas concerned are given in Gauss system of units.

III. RESULTS AND DISCUSSION

Figure 1 depicts the temperature dependence of magnetization $[M(T)]$ for MnSi under zero-field-cooling (ZFC) and field-cooling (FC) under an applied magnetic field $H = 0.1$ kOe, where the inset shows that for $H = 2$ kOe. The $M(T)$ curves for 0.1 kOe indicates a magnetic ordering transition at T_C, which corresponds to the well-known paramagnetic-helimagnetic (PM-HM) transition in MnSi [32, 33]. The transition temperature $T_C \approx 31$ K is determined from the dM/dT curve (the right axis in the inset). However, for $M(T)$ for $H = 2$ kOe, the magnetic transition is attributed to ordering from a paramagnetic to

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conical spin state, where the helimagnetic ordering is polarized into conical ordering by the external magnetic field [7].

In order to investigate the magnetic entropy change, the initial isothermal magnetization $M(H)$ around T_C for MnSi was measured, as shown in Fig. 2 (a). For the $M(H)$ curve far below T_C, it increases linearly with the increase of H below 5.6 kOe, and become almostly independent of H when exceeding 5.6 kOe. This result indicates the spin ordering behavior in this system. Below T_C, the spins are polarized gradually when H below 5.6 kOe, and becomes completely polarized when field exceeding 5.6 kOe. As it is well known, the magnetic entropy change (ΔS_M) can be calculated as [34]:

$$\Delta S_M(T, H) = \int_0^H \left(\frac{\partial S(T, H)}{\partial H} \right)_T dH \tag{1}$$

With Maxwell's relation: $[\partial S(T, H)/\partial H]_T = [\partial M(T, H)/\partial T]_H$ [35], $\Delta S_M(T, H)$ is written as:

$$\Delta S_M(T, H) = \int_0^H \left(\frac{\partial M(T, H)}{\partial T} \right)_H dH \tag{2}$$

Subsequently, ΔS_M can be calculated from experimental data as [36]:

$$|\Delta S_M| = \sum_i \left[\frac{(M_i - M_{i+1})_{H_i}}{T_{i+1} - T_i} \right] \Delta H_i \tag{3}$$

where M_i and M_{i+1} are the experimental data of the magnetization at T_i and T_{i+1} under a magnetic field H_i. Figure 2 (b) shows the $\Delta S_M(T, H)$ around T_C, which is obtained from the isothermal $M(H)$ curves. It can be seen that all curves almost exhibit a unimodal shape. However, there is an anomaly at ~ 28.5 K for the ΔS_M just below T_C. The inset of Fig. 2 (b) gives typical $\Delta S_M(T)$ curves for $H = 1.2$ and 1.8 kOe, both of which exhibit small valleys at ~ 28.5 K. This valley on $\Delta S_M(T)$ curve exists in the field range from ~ 1 kOe to ~ 2 kOe, and disappears gradually when the applied field exceeds 2 kOe. As it is known, MnSi exhibits a skyrmion transition at ~ 28.5 K in the field range from ~ 1.55 kOe to ~ 2.3 kOe [23]. The skyrmion transition should result into a change of magnetic entropy due to the ordering of the spins. Therefore, the valley on $\Delta S_M(T)$ curve should be attributed to the entropy change of skyrmion transition. Actually, a peak caused by the skyrmion lattice was found in the specific heat measurement, which confirms the magnetic entropy change of the skyrmion phase observed here [23].

The $\Delta S_M(T, H)$ can be scaled onto a phenomenological universal curve based on scaling relation [26, 27, 28]. According to scaling laws, the experimental $\Delta S_M(T, H)$ should

collapse onto two independent universal curves below and above T_C with proper reference temperatures and normalization. The $\Delta S_M(T)$ curves for different H were normalized as $|\Delta S_M(T)/\Delta S_M^{max}(T)|$ on the longitudinal coordinate, where the maximum of all curves are located on single point on the new normalized longitudinal coordinate. The horizontal coordinate is marked as the rescaled temperature θ, which is defined as [28]:

$$\theta = \frac{T - T_C}{T_r - T_C} \tag{4}$$

where T_r is the reference temperature. By choosing proper T_r, the equivalent points of the different experimental curves should collapse onto the same point on the universal curve [28]. The experimental $\Delta S_M(T, H)$ should collapse onto two independent universal curves below and above T_C respectively. The combination of the two universal curves below and above T_C just form a whole universal magnetic entropy change curve. After scaling and normalizing, the universal curve would be to impose the position of T_r at a value of $\theta = 1$.

Theoretical investigations have demonstrated that it is not necessary to use two reference temperatures below and above T_C but just a single one for a single magnetic phase material. However, in the case of a large demagnetization factor or the existence of multiple magnetic phases, the use of two reference temperatures is necessary. In this case, it is noticed that $\Delta S_M(T)$ for different H present asymmetry. In present experiment, the demagnetization energy has been carefully excluded as aforementioned. However, asymmetry of $\Delta S_M(T)$ still appears, which may be caused by the multiple magnetic phase transitions and magnetic fluctuation [37, 38, 39]. Therefore, two reference temperatures should be used, which are defined as [28]:

$$\theta = \begin{cases} \theta_- = (T_C - T)/(T_{r1} - T_C), T \leq T_C \\ \theta_+ = (T - T_C)/(T_{r2} - T_C), T > T_C \end{cases} \tag{5}$$

where T_{r1} and T_{r2} are the reference temperatures below and above T_C, respectively. Generally, temperatures at the full-width-at-half-maximum (FWHM) are chosen as the T_{r1} and T_{r2}, where T_{r1} corresponds to the one below T_C while T_{r2} to the one above T_C [29]. Then, in the new scaled coordinate systems, $\theta = \pm 1$ just corresponds to the positions of full-width-at-half-maximum (i.e. T_{r1} and T_{r2}).

The skyrmion phase has been absolutely suppressed by the external magnetic field of ~ 2.3 kOe. Thus, there is no skyrmion magnetic entropy change for $\Delta S_M(T, H)$ for a field exceeding 2.3 kOe. The scaling of $\Delta S_M(T, H)$ for higher fields exceeding 2.3 kOe

can generate an universal curve, which is independent of T and H. The phenomenological construction of the universal scaling of the experimental $\Delta S_M(T, H)$ is plotted in Fig. 3. The ΔS_M is normalized as $|\Delta S_M/\Delta S_M^{max}|$, and the temperature is plotted as a rescaled temperature θ. One can see that all experimental data collapse onto a single universal curve, except a small dispersion just below T_C which may be due to the strong magnetic fluctuation approaching T_C. Based on the universal scaling, an average curve is obtained, as shown as the red solid curve in Fig. 3. Obviously, the obtained average curve is universal and independent of the external measurement conditions, which is just determined by the intrinsic magnetization. According to the obtained universal curve, $\Delta S_M(T, H)$ at any T and H can be calculated. The inset of Fig. 3 shows the comparison of the calculated and experimental curves for the typical $\Delta S_M(T)$ for $H = 1.2$ kOe, where the calculated result agrees well with the experimental one except for that around 28.5 K.

This universal curve can be extrapolated to low fields to yield a calculated $\Delta S_M^{cal}(T, H)$. However, the entropy change of the skyrmion phase is not included by this calculation method. Therefore, through the comparison of the calculated ΔS_M and the experimental curve, the skyrmion magnetic entropy change can be derived by subtracting the $\Delta S_M^{cal}(T, H)$ from the experimental $\Delta S_M^{exp}(T, H)$. It can be seen that the calculated curve fits the experimental one very well except for the temperature range around 28.5 K. The discrepancy is attributed to the magnetic entropy caused by the skyrmion phase transition. Subsequently, the magnetic entropy change of the skyrmion (ΔS_M^{skyX}) can be obtained as:

$$|\Delta S_M^{skyX}(T)| = |\Delta S_M^{exp}(T) - \Delta S_M^{cal}(T)| \tag{6}$$

Figure 4 (a) shows the obtained $|\Delta S_M^{skyX}(T)|$ for different H. It can be seen that $|\Delta S_M^{skyX}(T)|$ reaches the maximum at $\sim$ 28.3 K. Figure 4 (b) plots the maximum of $|\Delta S_M^{skyX}(T)|$ vs H, which exhibits that the maximum of the $|\Delta S_M^{skyX}(T)|$ firstly increases with H, and subsequently decreases with H. The ΔS_M^{skyX} reaches the maximum at $H \approx 1.6$ kOe. As it is known, ΔS_M^{skyX} originates from the magnetic entropy change of the skyrmion phase transition. Thus, the maximum of the ΔS_M^{skyX} corresponds to the most drastic point of the skyrmion transition. Therefore, the results of the magnetic entropy change indicate that the maximal number of skyrmion vortices in MnSi is reached at $H \sim 1.6$ kOe, which is in agreement with the results of specific heat and detailed magnetic measurements [7, 23].

IV. CONCLUSION

In summary, the magnetic entropy change ΔS_M of MnSi has been investigated by the scaling analysis. The $\Delta S_M(T)$ curves in the field range from ~ 1 kOe to ~ 2 kOe exhibit small valleys at ~ 28.5 K, which correspond to the magnetic change caused by the skyrmion phase transition. The $\Delta S_M(T, H)$ curves in the higher field range collapse onto an universal curve after scaling, which is independent of T and H. Subsequently, magnetic entropy change of the skyrmion phase $[\Delta S_M^{skX}(T, H)]$ can be derived by subtraction of the calculated $\Delta S_M^{cal}(T, H)$ from the experimental $\Delta S_M^{exp}(T, H)$. It is demonstrated that the ΔS_M^{skX} reaches the maximum at ~ 1.6 kOe, which indicates that the maximal number of skyrmion vortices in MnSi is reached at $H \sim 1.6$ kOe.

V. ACKNOWLEDGEMENTS

[1] U. K. Roβler, A. N. Bogdanov, C. Pfleiderer, Nature (London) **442** (2006) 797.

[2] S. Muhlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, P. Boni, Science **323** (2009) 915.

[3] N. Nagaosa and Y. Tokura, Nat. Nanotechnol. **8** (2013) 899.

[4] F. Jonietz, S. Muhlbauer, C. Pfleiderer, A. Neubauer, W. Munzer, A. Bauer, T. Adams, R. Georgii, P. Boni, R. A. Duine, K. Everschor, M. Garst, A. Rosch, Science **330** (2010) 1648.

[5] X. Z. Yu, N. Kanazawa, W. Z. Zhang, T. Nagai, T. Hara, K. Kimoto, Y. Matsui, Y. Onose, Y. Tokura, Nat. Commun. **3** (2012) 988.

[6] A. Fert, V. Cros, J. Sampaio, Nat. Nanotechnol. **8** (2013) 152.

[7] A. Bauer and C. Pfleiderer, Phys. Rev. B **85** (2012) 214418.

[8] X. Z. Yu, N. Kanazawa, Y. Onose, K. Kimoto, W. Z. Zhang, S. Ishiwata, Y. Matsui, Y. Tokura, Nat. Mater. **10** (2011) 106.

[9] A. Tonomura, X. Yu, K. Yanagisawa, T. Matsuda, Y. Onose, N. Kanazawa, H. S. Park, Y. Tokura, Nano Lett. **12** (2012) 1673.

[10] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, Y. Tokura, Nature (London) **465** (2010) 901.

[11] W. Munzer, A. Neubauer, T. Adams, S. Mhlbauer, C. Franz, F. Jonietz, R. Georgii, P. Boni, B. Pedersen, M. Schmidt, A. Rosch, C. Pfleiderer1, Phys. Rev. B **81** (2010) 041203(R).

[12] J. S. White, K. Prsa, P. Huang, A. A. Omrani, I. Zivkovic, M. Bartkowiak, H. Berger, A. Magrez, J. L. Gavilano, G. Nagy, J. Zang, H. M. Ronnow, Phys. Rev. Lett. **113** (2014) 107203.

[13] T. Adams, A. Chacon, M. Wagner, A. Bauer, G. Brandl, B. Pedersen, H. Berger, P. Lemmens, C. Pfleiderer, Phys. Rev. Lett. **108** (2012) 237204.

[14] N. Manyala, Y. Sidis, J. F. DiTusa, G. Aeppli, D. P. Young, Z. Fisk, Nature (London) **404** (2000) 581.

[15] C. Pfleiderer, S. R. Julian, G. G. Lonzarich, Nature (London) **414** (2001) 427.

[16] H. Watanabe, S. A. Parameswaran, S. Raghu, A. Vishwanath, Phys. Rev. B **90** (2014) 045145.

[17] C. Pfleiderer, G. J. McMullan, S. R. Julian, G. G. Lonzarich, Phys. Rev. B **55** (1997) 8330.

[18] P. Bak and M. H. Jensen, J. Phys. C: Solid St. Phys. **13** (1980) L881.

[19] S. A. Brazovskii, Sov. Phys. JETP **41** (1975) 85.

[20] K. Huang, *Statistical Mechanics*, 2nd ed. (Wiley, New York, 1987).

[21] S. Buhrandt and L. Fritz, Phys. Rev. B **88** (2013) 195137.

[22] L. Zhang, D. Menzel, C. M. Jin, H. F Du, M. Ge, C. J. Zhang, L. Pi, M. L. Tian, Y. H. Zhang, Phys. Rev. B **91** (2015) 024403.

[23] A. Bauer, M. Garst, C. Pfleiderer, Phys. Rev. Lett. **110** (2013) 177207.

[24] H. F. Du, W. Ning, M. L. Tian, Y. H. Zhang, Phys. Rev. B **87** (2013) 014401.

[25] H. F. Du, J.. DeGrave, F. Xue, D. Liang, W. Ning, J. Y. Yang, M. L. Tian, Y. H. Zhang, S. Jin, NanoLett. **14** (2014) 2026.

[26] V. Franco, J. S. Blazquez, A. Conde, Appl. Phys. Lett. **89** (2006) 222512.

[27] V. Franco, A. Conde, J. M. Romero-Enrique, J. S. Blazquez, J. Phys.: Condens. Matter **20** (2008) 285207.

[28] V. Franco, A. Conde, Int. J. Refrigerat. **33** (2010) 465.

[29] L. Zhang, J. Y. Fan, W. Tong, L. S. Ling, L. Pi, Y. H. Zhang, Physica B **407** (2012) 3543.

8

[30] V. A. Dyadkin, S. V. Grigoriev, D. Menzel, D. Chernyshov, V. Dmitriev, J. Schoenes, S. V. Maleyev, E. V. Moskvin, H. Eckerlebe, Phys. Rev. B **84** (2011) 014435.

[31] A. K. Pramanik and A. Banerjee, Phys. Rev. B **79** (2009) 214426.

[32] M. Janoschek, M. Garst, A. Bauer, P. Krautscheid, R. Georgii, P. Boni, C. Pfleiderer, Phys. Rev. B **87** (2013) 134407.

[33] J. Kindervater, W. Hauβler, M. Janoschek, C. Pfleiderer, P. Boni, M. Garst, Phys. Rev. B **89** (2014) 180408(R).

[34] V. Pecharsky, K. Gscheidner, J. Magn. Magn. Mater. **200** (1999) 44.

[35] A. H. Morrish, The Physical Principles of Magnetism, Wiley, New York, 1965 (Chapter 3).

[36] W. J. Lu, X. Luo, C. Y. Hao, W. H. Song, Y. P. Sun, J. Appl. Phys. **104** (2008) 113908.

[37] C. Pappas, E. Lelievre-Berna, P. Falus, P. M. Bentley, E. Moskvin, S. Grigoriev, P. Fouquet, B. Farago, Phys. Rev. Lett. **102** 197202 (2009).

[38] S. V. Grigoriev, N. M. Potapova, S. A. Siegfried, V. A. Dyadkin, E.V. Moskvin, V. Dmitriev, D. Menzel, C. D. Dewhurst, D. Chernyshov, R. A. Sadykov, L. N. Fomicheva, A.V. Tsvyashchenko, Phys. Rev. Lett. **110** (2013) 207201.

[39] S. V. Grigoriev, S. V. Maleyev, E. V. Moskvin, V. A. Dyadkin, P. Fouquet, H. Eckerlebe, Phys. Rev. B **81** (2010) 144413.
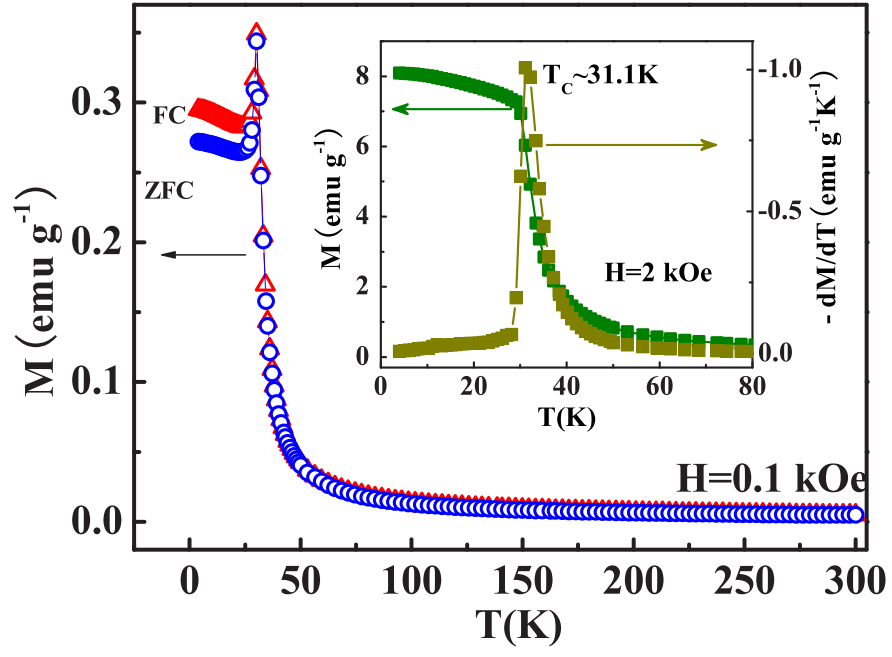
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FIG. 1: (Color online) Temperature dependence of the magnetization $[M(T)]$ for MnSi for $H = 0.1$ kOe [the inset shows $M(T)$ (left axis) and dM/dT (right one) for $H = 2$ kOe].
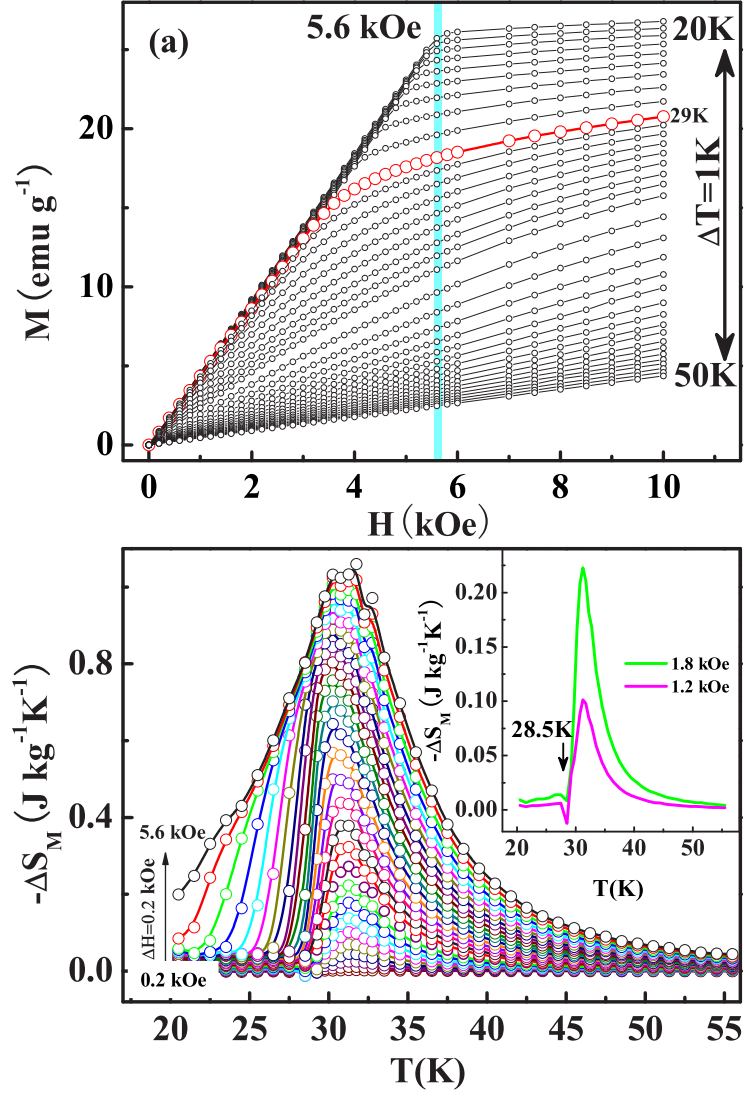
FIG. 2: (Color online) (a) Initial magnetization $M(H)$ around T_C for MnSi (the red curve corresponds that at 29 K); (b) magnetic entropy change (ΔS_M) around T_C for different fields (the inset shows the typical curves for $H = 1.2$ and 1.8 kOe).
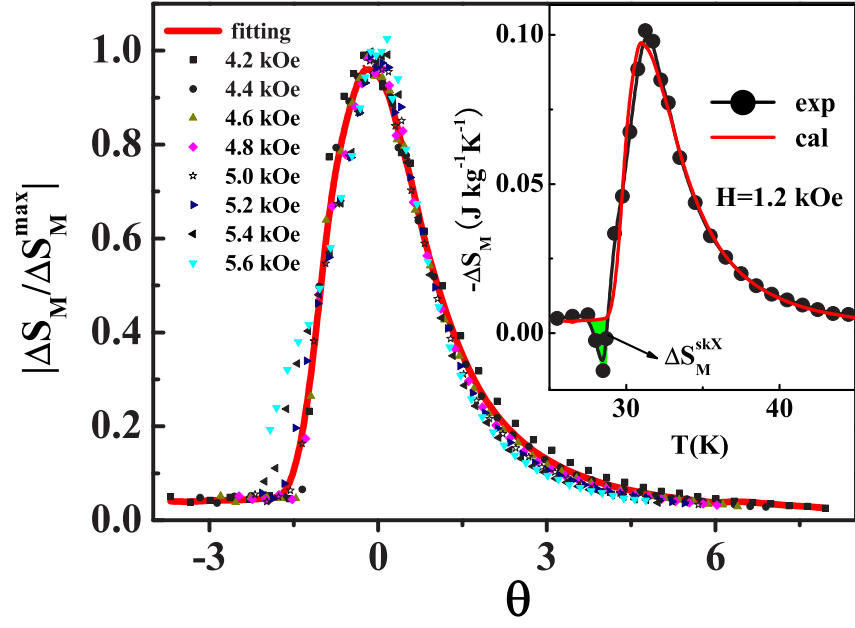
FIG. 3: (Color online) Scaling of $\Delta S_M(T, H)$ curves for MnSi (the inset shows the comparison of the calculated and the experimental curve for $H = 1.2$ kOe).
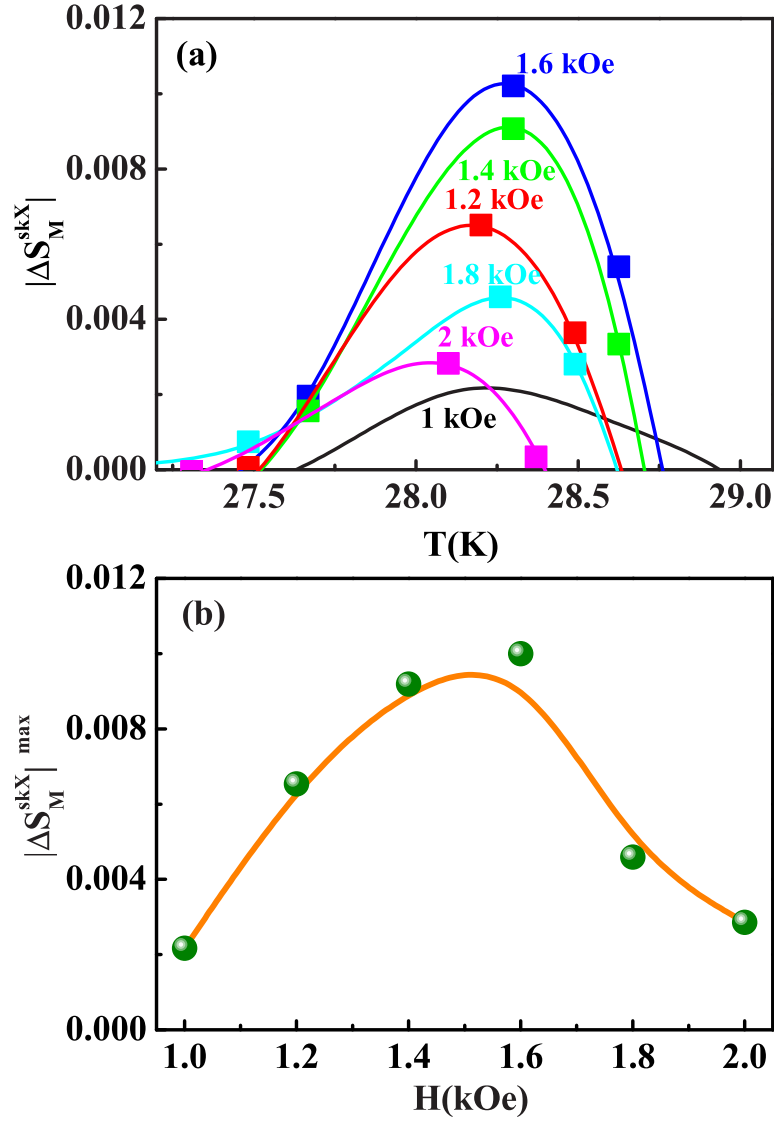
FIG. 4: (Color online) (a) Magnetic entropy change of skyrmion transition $[|\Delta S_M^{skyX}(T)|]$ for different fields as a function of temperature; (b) maximum of the $|\Delta S_M^{skyX}(T)|$ vs H (the curves are guided on eye).